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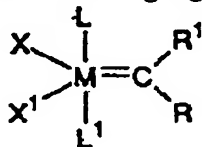
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## CLAIMS

## [Claim(s)]

1. It is the Manufacture Approach of Poly Cycloolefin Which Comes to Contain Process Which Carries Out Polymerization of the Olefin under Existence of Cross Linking Agent and Catalyst, and Said Cross Linking Agent



Contains Peroxide, and Said Catalyst is Formula. :

inside of formula, and M -- Os or Ru -- it is -- R and R1 -- hydrogen or C1-C -- 20 alkyl C2-C20 alkenyl, C2-C20 alkynyl, C2-C20 alkoxy carbonyl, Aryl, C1-C20 carboxylate, and C1-C20 alkoxy \*\* It is chosen independently of the hydrocarbon chosen from the group which consists of C2-C20 alkenyloxy, C2-C20 alkynyloxy, and aryloxy. X and X1 are chosen independently of all anion nature ligands -- having -- and -- L and L1 are chosen independently of all neutral electronic donors.

The manufacture approach of the poly cycloolefin which is a \*\* ruthenium or an osmium carbene complex.

2. Approach containing peroxide chosen from group which said cross linking agent becomes from t-butyl peroxide, 2, 5-dimethyl -2, 5-G (tert-butylperoxy) hexyne -3, G tert-butyl peroxide and 2, 5-dimethyl -2, 5-G (tert-butylperoxy) hexanes, or such mixture according to claim 1.

3. the amount of a cross linking agent -- about [ of the mixture of a catalyst, a cross linking agent, and a monomer ] -- about [ 0.01 v% - ] -- the approach according to claim 1 of being 10 v%.

4. Approach according to claim 1 olefin is annular or non-ring type.

5. Approach according to claim 1 L and L1 are trialkylphosphine ligands.

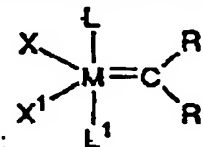
6. Approach according to claim 5 by which L and L1 are chosen independently of group which consists of P (isopropyl)3, P (cyclopentyl)3, and P (cyclohexyl)3.

7. Approach according to claim 1 polymerization happens under [ of a solvent ] absent.

8. Approach according to claim 4 annular olefin is norbornene or norbornene derivative.

9. Approach according to claim 8 annular olefin is dicyclopentadiene.

10.a) the process which carries out the polymerization of the olefin with a catalyst under existence of a



peroxide, and generates the poly cycloolefin polymer -- [ -- said catalyst -- formula:

inside of formula, and M -- Os or Ru -- it is -- R and R1 -- hydrogen or C1-C -- 20 alkyl

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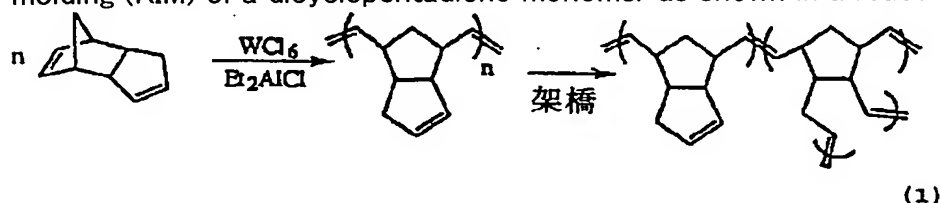
## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

Field of peroxide bridge formation invention of a ROMP polymer This invention relates to the manufacture approach of a polymer of having high density bridge formation. Furthermore, this invention relates to the olefin double decomposition polymerization method which generates the polymer which has high density bridge formation in detail.

Background The double decomposition polymerization of an annular olefin brings about a high polymer by the ring breakage double decomposition polymerization (ROMP). The initial catalyst used for ROMP was the thing of a Ziegler type. These catalysts are not defined so clearly and generated by the reaction of the reducing agent like initial transition-metals halide and alkylaluminum.

The double decomposition polymerization developed for the purpose of marketing is the reaction injection molding (RIM) of a dicyclopentadiene monomer as shown in a reaction 1.



By this approach, a metal halide component and a reducing-agent component are separately mixed with a monomer. When it mixes, a catalyst is generated and mixture is poured into the metal mold for hardening. This kind of polymerization was indicated as the various catalysts which use a tungsten or molybdenum as the base were indicated by U.S. Pat. No. 4,400,340, 4,380,617, and the 4,701,510 specification. The solvent resistance to which the polymer obtained increased, for example, low gel swelling, is acquired by addition of the cross linking agent of a low. However, typically, at these reactions, a monomer needs to be refined by altitude and water and the catalyst poison like alcohol need to be avoided.

The initial transition metal catalyst for ROMP of an annular olefin specified well was developed over the past eight years. as an instantiation catalyst -- L.K -- Johnson et al., J.Am.Chem.Soc., 115, 8167-8177 (1993) and D.L.Gin et al., J.Am.Chem.Soc., 114, and the thing indicated by 3167-3169 (1992) are mentioned. These catalysts are easy organometallic complexes which include the metal carbon to carbon bond of a large number which carry out the polymerization of the olefin by the "living" approach in almost all cases. It was used for mono dispersion of these catalysts being carried out mostly, and preparing a uniform polymer on structure. for example, R.H.Grubbs and Pure Appl.Chem. -- the United States patent application 08th for which it applied on July 29, 1994 transferred to the patentee of A31 (11), 1829-1833 (1994), and this invention / No. 282,827 -- and -- said -- refer to the 08th / No. 282,826 (these all are contained in this specification as reference). Although such a catalyst studied first used a tungsten and molybdenum as the base, the VIII group ROMP catalyst of activity series was prepared recently. These new catalysts enable wide range functionality to be crowded for a polymer side chain. The most important \*\* of the complex of this family is a BISUHOSEFIN dihalo ruthenium carbene complex. For example, refer to S.T.Nguyen et al. and J.Am.Chem.Soc. which are contained in this specification as it is as reference, 115, 9858-9859 (1989), and a U.S. Pat. No. 5,312,940 specification. Moreover, it can be used for these catalysts of these compounding fine chemicals and a block copolymer. For example, G.C.Fu et al., J.Am.Chem.Soc., 115, 9856-9857 (1993), Z.Wu et al., Macromolecules, 26, 4975-4977 (1993) Z.Wu et al., Macromolecules, 27, 6700-6703 (1994), and L.K -- refer to Johnson et al., J.Am.Chem.Soc., 115, 8130-8145 (1993), S.T.Nguyen et al., J.Am.Chem.Soc., 114, 3974-3975 (1992) and B.T.Flatt et al., Organometallics, 13, and 2728-2732 (1994).

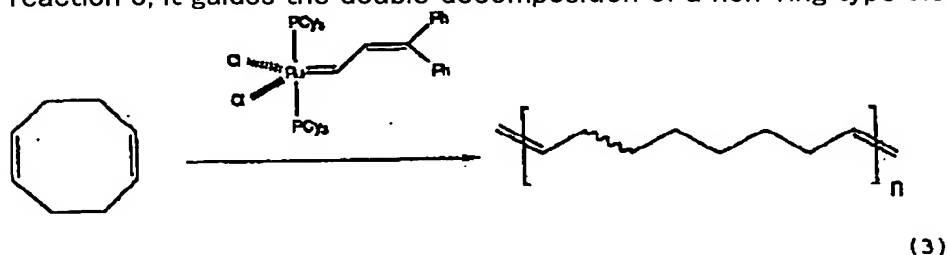
The ruthenium double decomposition catalyst defined clearly first was prepared by the reaction of tris (triphenyl

phosphine) ruthenium dichloride and 1,1-diphenyl cyclopropene, as shown in a reaction 2.



The diazomethane derivative prepared by the approach of the compound which uses other ruthenium analogs as the base, for example, a phosphorus-containing (Wittig mold) reagent, and others catalyzes a double decomposition reaction again.

The above-mentioned ruthenium carbene complex catalyzes the living polymerization of norbornene or the other distorted olefins existing (strain-ed), for example, transformer-cyclooctane. An end cap is carried out and the norbornene polymer obtained shows the polydispersity of 1.2. The width of face of polydispersity is as a result of late initiation of the catalyst about the rate of a polymerization. Easy exchange of the triphenyl phosphine ligand brought about remarkable increase of activity about tri-cyclohexyl phosphine. A tri-cyclohexyl phosphine ruthenium carbene catalyst carries out the polymerization of the distorted olefin which is not, and as shown in a reaction 3, it guides the double decomposition of a non-ring type olefin.



Application of the norbornene in an aqueous solvent or a nonaqueous nature solvent and a norbornene Mr. derivative, for example, the tri-cyclohexyl phosphine ruthenium (or osmium) carbene catalyst in the polymerization of a dicyclopentadiene (DCPD), is indicated by the U.S. Pat. No. 5,342,909 specification contained in this specification as reference again.

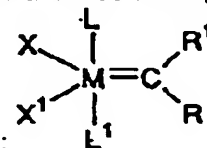
The polymer which has high density bridge formation is desirable because of those improved mechanical strength and low gel swelling. Bridge formation is often attained by use of the compound which reacts with a polymer and forms bridge formation. For the ease of processing, a cross linking agent is often added in a reaction mixture including a monomer and a catalyst. When these matter changes or controls a polymerization reaction or it poisons a catalyst, other means need to be found out although bridge formation is attained.

Epitome of invention This invention cancels these faults by making the contact double decomposition polymerization of an olefin possible under existence of a modifier. It can manufacture simply under existence of a modifier, and efficiently without the polymer which has very high crosslinking density deactivating a catalyst. The polymer obtained is solvent resistance and mechanically strong. Moreover, the monomer of a certain kind which has the low purity which can carry out a polymerization to a success flesh side by the usual approach (for example, 85 - 95% of dicyclopentadiene)

According to this invention, a polymerization is carried out using the double decomposition catalyst reported to \*\*\*\*, and a viaduct consistency ingredient can be manufactured.

Generally, by the approach of this invention, a "modifier" or a cross linking agent is mixed with a monomer, for example, an annular olefin, and a catalyst. A monomer carries out a polymerization according to a double decomposition mechanism, and then a cross linking agent generates the active species which reacts with the polymer obtained by decomposing at an elevated temperature, and forms bridge formation. Under existence of a modifier, deactivation of the catalyst is not carried out or it is not poisoned.

It sets like 1 operative condition, and this invention is the manufacture approach of the poly cycloolefin which comes to contain the process which mixes an olefin with a catalyst under existence of a cross linking agent



peroxide, and the cross linking agent contains a peroxide, and a catalyst is a formula. :

( M being Os or Ru among a formula.) R and R1 Hydrogen or C1-C20 alkyl, C2-C20 alkenyl, C2-C20 alkynyl, C2-

C20 alkoxy carbonyl, aryl, C1-C20 carboxylate, C1-C20 alkoxy \*\*C2-C20 alkenyloxy, it chooses independently of the hydrocarbon chosen from the group which consists of C2-C20 alkenyloxy and aryloxy -- having -- X and X1 are chosen independently of all anion nature ligands -- having -- and -- L and L1 are chosen independently of all neutral electronic donors.

They are a \*\* ruthenium or an osmium carbene complex.

another operative condition -- it is the manufacture approach of the poly cycloolefin which comes to contain the process which sets like, this invention carries out the polymerization of the olefin with a catalyst under existence of the above-mentioned peroxide, disassembles the peroxide in a polymer, and generates the reactant kind of a peroxide, and forms bridge formation into a polymer using the reactant kind.

In either of these embodiments, the polymer generated by the double decomposition reaction can generate the polymer which postcure is carried out at an elevated temperature and has high density bridge formation.

As a suitable monomer, the monomer which can carry out a polymerization by the double decomposition reaction, for example, an annular olefin, (a 2 ring type diolefin and a 3 ring type diolefin are included in a monocycle type olefin list), a non-ring type olefin, for example, the diolefin of a non-ring type, or a high-class olefin is mentioned.

The catalyst as which L and L1 are chosen as a desirable catalyst independently of P (isopropyl)3, P (cyclopentyl)3, and P (cyclohexyl)3 is mentioned to the approach.

A desirable cross linking agent is a peroxide, for example, alkyl peroxide, especially tert-butyl peroxide or G t-butyl peroxide, 2, the 5-dimethyl -2, and 5-G (tert-butylperoxy) hexyne. - They are 3, 2, the 5-dimethyl -2, 5-G (tert-butylperoxy) hexanes, or such mixture. As for the amount of the peroxide in a reaction mixture, it is desirable that they are 0.01 v% - of total monomer / catalyst / cross linking agent mixture, and 10 v%.

A polymerization can be performed under [ of a solvent ] absent.

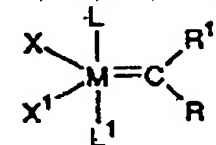
Detailed explanation Generally, the viscosity of mixture increases, and the approach of this invention needs the thing of the pour point for which a catalyst, a monomer, and a modifier are mixed together at a room temperature until it turns down exactly. Next, the mixture is poured into the metal mold by which the preheating was preferably carried out to about 35 to 45 degree C - about 140 degrees C.

Generally the time amount to generation of heat of a polymerization reaction is about 5 minutes - 6 hours, and the viscosity of mixture increases quickly at the time.

The time amount to generation of heat under double decomposition is determined by the ratio of the monomer pair catalyst in mixture, and the purity of a monomer also in other factors. The ratios of a monomer pair catalyst may be about 100:1 - abbreviation 10,000:1. The ratio about a specific monomer and a specific catalyst may be out of this range, in order to control the final property of the polymer manufactured. At about 100:1 ratio, there is an inclination for a polymer to be hard comparatively and, on the other hand, there is an inclination for the polymer obtained to be elasticity, by the ratio of about 10,000:1. It is expected that a ratio higher than 10,000:1 produces a further elastic polymer.

Immediately after generation of heat, a polymer is removed from metal mold and then postcure is carried out over a period almost equal to an elevated temperature and one to 3 of a peroxide [ in / preferably / in a top / decomposition point / of a peroxide / the temperature ] half-life. High density bridge formation is attained in a polymer in postcure.

As a catalyst suitable for using it for the approach of this invention, it is stable under existence of various functional groups, and the ruthenium or osmium carbene complex which can be used for catalyzing the olefin double decomposition reaction of an annular olefin without distortion and some non-ring type olefins is mentioned. These catalysts and those manufacture approaches are indicated by U.S. Pat. No. 5,342,909 and 5,312,940, and these each is contained in this specification as reference. Generally, these catalysts are formulas.



( M being Os or Ru among a formula, and R and R1 becoming independent of the hydrocarbon chosen from the group which consists of hydrogen or C1-C20 alkyl, C2-C20 alkenyl, C2-C20 alkynyl, C2-C20 alkoxy carbonyl, aryl, C1-C20 carboxylate, C1-C20 alkoxy \*\*C2-C20 alkenyloxy, C2-C20 alkynyloxy, and aryloxy.) it chooses -- having -- X and X1 are chosen independently of all anion nature ligands -- having -- and -- L and L1 are chosen independently of all neutral electronic donors.

It is alike and can indicate more. It may join together together as occasion demands, and any two, X, X1, L, and L1, or three may form a chelation multidentate ligand.

As instantiation of L and L1, a phosphine, a sulfonation phosphine, phosphite, phosphinite, a phospho nit, an arsine, a stibine, the ether, an amine, an amide, a sulfoxide, carbonyl, nitrosyl, a pyridine, or a thioether is mentioned.

In the desirable embodiment, the double decomposition polymerization of this invention is performed using the catalyst whose at least one of the alkyl groups of a phosphine as shown in the ruthenium of the above-mentioned formula L and whose L1 are trialkylphosphine or an osmium carbene catalyst especially the United States patent application 08th / No. 282,827, and this/[ 08th ] No. 282,826 specification is the second class alkyl group or a cycloalkyl radical, for example, a cyclopentyl group, a cyclohexyl radical, or an isopropyl group. It may be desirable to protect some catalysts used for this invention from an ambient atmosphere, for example, oxygen. For example,  $2(P(\text{cyclopentyl})_3)Cl_2RuCHCHPh_2$  and its cyclohexyl phosphine analog may be stored all over a desiccation box under nitrogen-gas-atmosphere until they are needed. The amount of a request of a catalyst is measured and capped by the vial all over a desiccation box, and it may be removed from the glove compartment for use under polymerization reaction.

The norbornene derivative which contains the olefin which can carry out a polymerization according to a double decomposition mechanism, for example, JISHIKURO Pentagin, as a suitable monomer is mentioned. A monomer may be annular or a non-ring type, and the latter is a diolefin at least, in order to carry out a polymerization. It may be desirable to pretreat a monomer before contact for a catalyst. For example, DCPD may be commercially supplied as 95% of purity. A p-tert-butyl catechol is an additive for preventing an early radical polymerization. A monomer may be refined before a double decomposition polymerization. Filtration of DCPD by the activated neutral aluminum oxide (Fluka #06300) removes an antioxidant.

Since the catalyst is stable under existence of a functional group, a monomer can be mixed with the modifier like a peroxide before a polymerization. The alkyl peroxide of 0.01–10v% of amount of a certain kind is especially typically useful (more amounts within the limits of this tend to fall polymerization degree). Such high crosslinking density is given to a polymer that the alkyl peroxide like t-butyl peroxide containing G t-butyl peroxide is desirable. Two sorts of desirable peroxides are marketed as the trade name LUPERSOL 130 containing 2, the 5-dimethyl -2, 5-JI (tert-butylperoxy) hexyne -3, and G t-butyl peroxide, and LUPERSOL 101 containing 2, the 5-dimethyl -2, 5-JI (tert-butylperoxy) hexane, and G tert-butyl peroxide (from ELS Atochem N-orth America Inc. Fine Chemicals Group).

By the approach of this invention, preferably, after generation of heat of a polymerization reaction, and in postcure, a peroxide decomposes at an elevated temperature and generates a free radical or other reactant kinds. The polymer obtained increases solvent resistance sharply and this is considered for the crosslinking density which increased.

The crosslinking density which increased has the gel swelling and the correlation which decreased. It turned out that it has the solvent resistance which increased remarkably compared with the polymer prepared without adding a peroxide, although the ruthenium catalyst with the polymer same as shown in Table I prepared according to this invention explained in more detail below was used.

Example 1: Purification of a DCPD monomer DCPD 500mL is filtered to a 1l. round bottom flask with the sintered glass filter of the middle porosity of 150mL(s) which contain a 1 inch aluminum oxide under a vacuum. Before use, an aluminum oxide is stored at 120 degrees C in oven. DCPD of low grade which has the low congealing point can be used after the same purification processing.

**Example 2: Degassing of a DCPD monomer** A 1 inch magnetic stirring rod is added to a round bottom flask including the above-mentioned purification DCPD. A flask is put into a 30–35-degree C water bath, and it stirs under the vacuum of 0.10mmHg for 1.5 hours. Next, deaerated DCPD is stored under a vacuum, it protects from light, and an early radical polymerization is prevented.

Example 3: DCPD polymerization which does not use a peroxide DCPD (147.9g, 150mL, 1.12 mols, 5000Eq) and (P(cyclopentyl) 3)  $2\text{Cl}_2\text{RuCHCHPh}_2$  (mg [ 188.5 ], 0.224 millimol, 1Eq) by which were refined as mentioned above by the Erlenmeyer flask of 250mL(s) containing a 1 inch magnetic stirring rod, and degassing was carried out to it were added.

Stirring was started and the argon was slowly slushed into the flask. The solution of the orange was stirred for 8 minutes at the room temperature under the argon ambient atmosphere until it became viscosity to altitude. Next, it poured into the crystallization pan (diameter of 14.6cm) which stored the solution at 120 degrees C beforehand, and exposed to air. The solution began to gel after 2 minutes, and generating of smoke was observed after 2 minutes. At this time, what the polymerization completed is clear and cooled the crystallization pan to the room temperature. The polymer was easily separated from the side face of a crystallization pan. Postcure of the polymer was carried out over 3 hours at 120 degrees C, and Pori (DCPD) (141.1g, 95.4% of yield) was obtained.

Example 4: DCPD polymerization which uses a peroxide DCPD (147.9g, 150mL, 1.12-mol, 5000Eq) and (P

(cyclopentyl) 3)  $2\text{Cl}_2\text{RuCHCHPh}_2$  catalyst (188.5mg, 0.224 millimol, 1Eq) by which was refined as mentioned above by the Erlenmeyer flask of 250mL(s) containing a 1 inch magnetic stirring rod, and degassing was carried out to it was added. Next, LUPERSOL-130 (4.5mL, 3%v/v) was added, and stirring was started on the basis of the late flow of an argon. The solution of the orange was stirred for 8 minutes at the room temperature under the argon ambient atmosphere until it became viscosity to altitude. Next, the solution was poured into the crystallization pan (diameter of 14.6cm) by which the preheating was carried out to 120 degrees C, and it exposed to air. The solution began to gel after 2 minutes, and generating of smoke was observed after 2 minutes. At this time, what the polymerization completed is clear and cooled the crystallization pan to the room temperature. It dissociated easily from the side face of a crystallization pan, and the polymer carried out postcure of this over 3 hours at 120 degrees C. Pori (DCPD) (139.4g, 94.3% of yield) was manufactured.

Example 5: Gel swelling of Pori (DCPD) The following table 1 summarizes the comparison result of a gel swelling experiment of Pori (DCPD) prepared according to the approach of the conventional technique and this invention.

It is the following, and Pori (DCPD) of this invention was made and prepared. it was refined as mentioned above by the Erlenmeyer flask of 250mL(s) containing 1 1/2 inch magnetic \*\*\*\*\*, and degassing was carried out to it - LUPERSOL 130 (0.68mL, 1%v/v) was DCPD(ed) and (67.1g, 68.1mL, 0.51 mols, 4000Eq) added continuously. The mixture was slowly stirred for 3 minutes on the basis of a nitrogen style.

(P(cyclopentyl) 3)  $2\text{Cl}_2\text{RuCHCHPh}_2$  (mg [ 106.0 ], 0.126 millimol, 1Eq) was added, and mixture was stirred. Stirring was continued under nitrogen-gas-atmosphere mind at the room temperature until mixture became viscosity to altitude after about 7 minutes and 30 seconds. Next, the solution was poured into the aluminum metal mold (2.54 cmx1.27 cmx20.32 cm die length) beforehand stored at 40 degrees C, and it exposed to air. After about 3 minutes, it was begun at the temperature of 157 degrees C to gel the solution by peak generation of heat for 14 minutes and 35 seconds. At this time, what the polymerization completed is clear and cooled metal mold to the room temperature. The polymer obtained was removed from aluminum metal mold, and postcure was carried out over 1 1 / 2 hours at 190 degrees C.

The gel swelling sample was measured, and it put into the flask which contains either an alt.-xylene or toluene as a solvent, and flowed back over several hours. After heating in a solvent over the period of predetermined time amount, the piece was removed, and it was made to dry and measured again. % gel swelling was calculated by breaking the weight augend by the initial mass, and hanging 100.

表 1

実験	モノマー／触媒	過酸化物質 % 容量	後硬化前の ゲル膨潤	後硬化後の ゲル膨潤	後硬化	ゲル膨潤 の条件	過酸化物質 の型
1	5000:1	0	-	155%	120°C/1½ h	B	E
2	5000:1	3%	-	116%	120°C/1½ h	B	E
3	5000:1	0	70.85%	-	-	A	
4	5000:1	0		67.28%	190°C/1½ h	A	
5	5000:1	0		62.49%	190°C/1½ h	A	
6	5000:1	3%	51.40%	-	-	A	E
7	5000:1	3%	-	0.35%	190°C/1½ h	A	E
8	5000:1	3%	-	0.51%	190°C/1½ h	A	E
9	5000:1	1%	65.45%	-	-	A	E
10	5000:1	1%	-	0%	190°C/1 h	A	E
11	7500:1	3%		反応せず <sup>1</sup>			H
12	5000:1	4%	-	1.78%	190°C/1 h	D	F
13	5000:1	4%	-	1.31%	190°C/1 h	D	F
14	7500:1	3%		反応せず <sup>1</sup>	-		I
15	7500:1	4%	-	1.60%	190°C/1 h	C	G
16	7500:1	4%	-	1.22%	190°C/1 h	C	G
17	5000:1	0.4%		反応せず <sup>1</sup>			J
18	5000:1	1%		反応せず <sup>1</sup>			K
19	5000:1	1%		不完全な 反応			L

A: by the visual inspection of one sample -- reflux -- carrying out -- alt.xylene B: reflux of 11 hours -- carrying out -- toluene C: reflux of 2 hours -- carrying out -- alt.xylene D: reflux of 8 hours -- carrying out -- alt.xylene E: LUPERSOL 130 F: t-butyl peroxide 98% G: LUPERSOL 101 H: benzoyl peroxide 97% I: LUCIDOL 75 (trade name of benzoyl peroxide marketed from Elf Atochem North America Inc.) of 10.5 hours

J: it sees in a t-amyl pel benzoate K: tert butylhydroperoxide L: LUPERSOL 231 table -- as -- gel swelling -- the peroxide (Mold E and G[-- respectively -- LUPERSOL 130 and a 101] list -- mold F [t-butyl peroxide]) of this invention of a certain kind

In Pori (DCPD) by which the polymerization was carried out under existence, it decreases remarkably. Addition of these peroxides of this invention and the technique of postcure bring about Pori (DCPD) which hardly shows gel swelling. In this way, resistance [ as opposed to solvent absorption in Pori (DCPD) manufactured according to this invention ] increases.

Moreover, the data about the trial which does not succeed in Table 1 carrying out the polymerization of the DCPD under existence of the peroxide of various others are also included (experiments 11, 14, 17, 18, and 19). It was shown clearly that a polymerization hardly happens or the visual inspection of these samples does not happen at all. It is thought that the polymerization was not attained for control of others of the catalyst poison by these peroxides or a polymerization reaction.

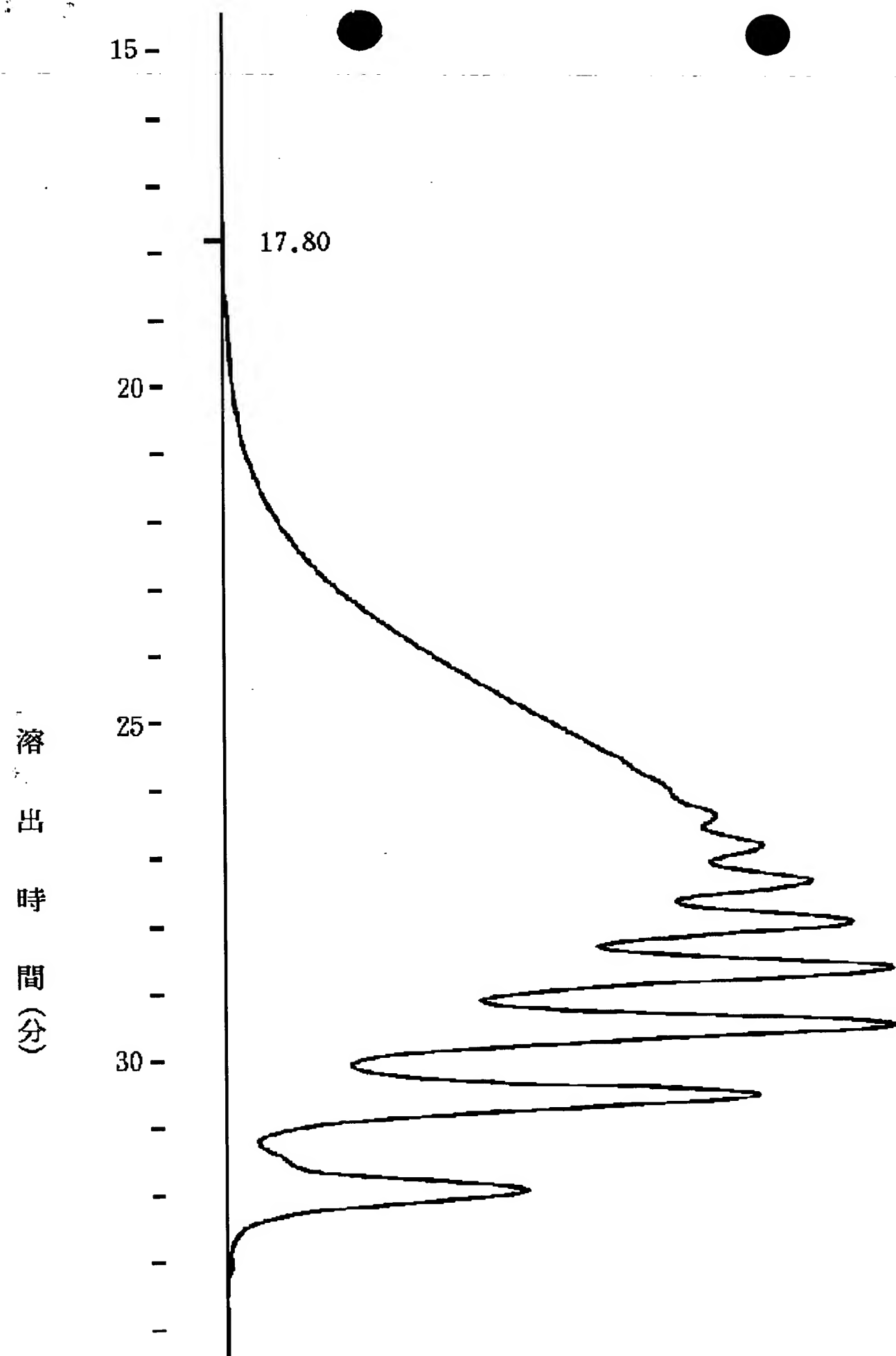
Moreover, a table also contains the data (experiments 1 and 2) which suggest that it is not effective in the postcure of 120 degrees C / 1.5 hours producing high density bridge formation rather than the postcure of 190 degrees C / 1 hour.

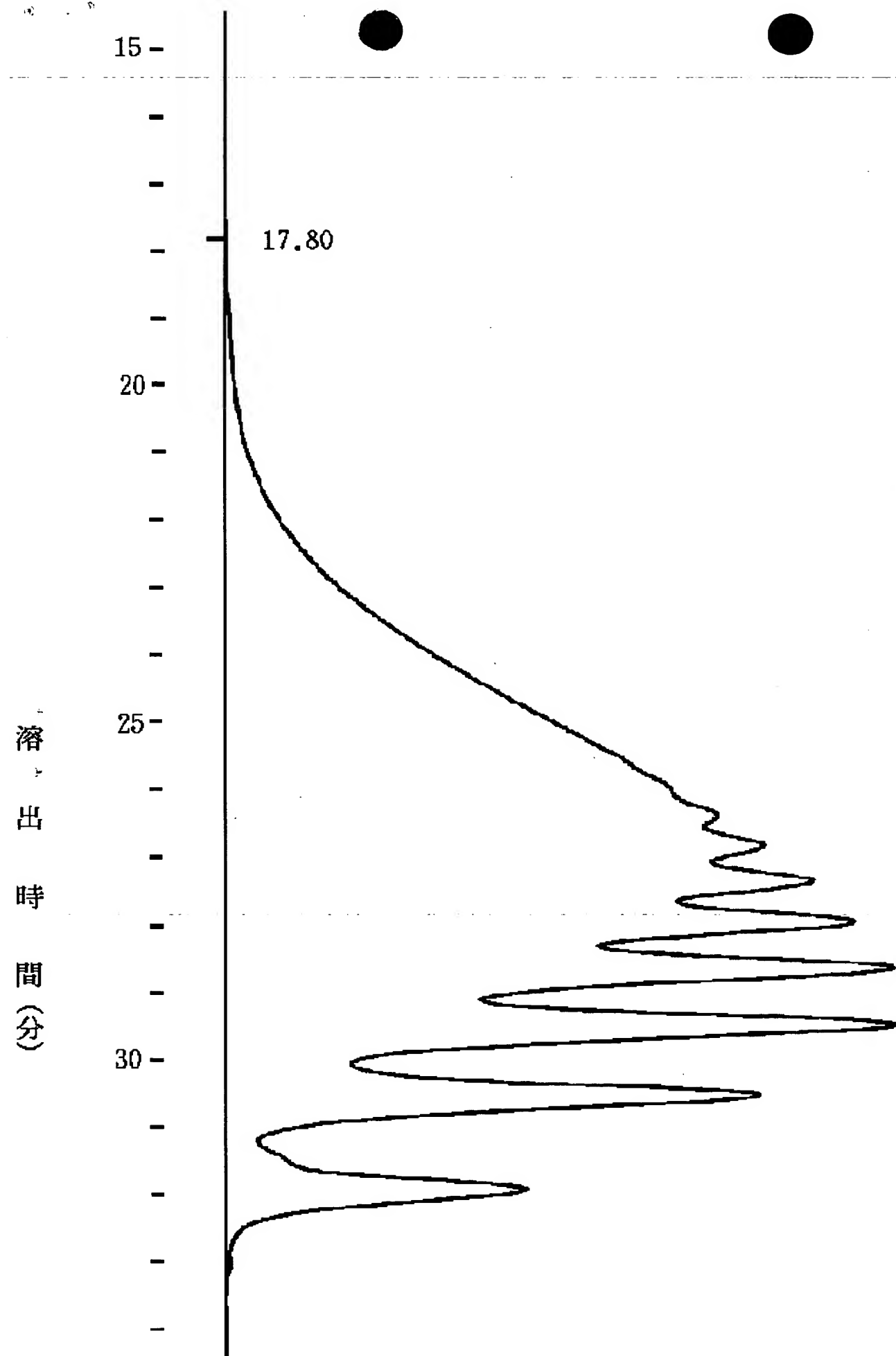
Although this invention is indicated considering the desirable embodiment and desirable example which are explained in full detail above as reference, it should be understood that these examples are meant as instantiation rather than the semantics of limitation. It is because it has the intention of what many amelioration within the range of this invention and pneuma thinks of easily to this contractor and has the intention of an attached claim including such amelioration, if it becomes what.

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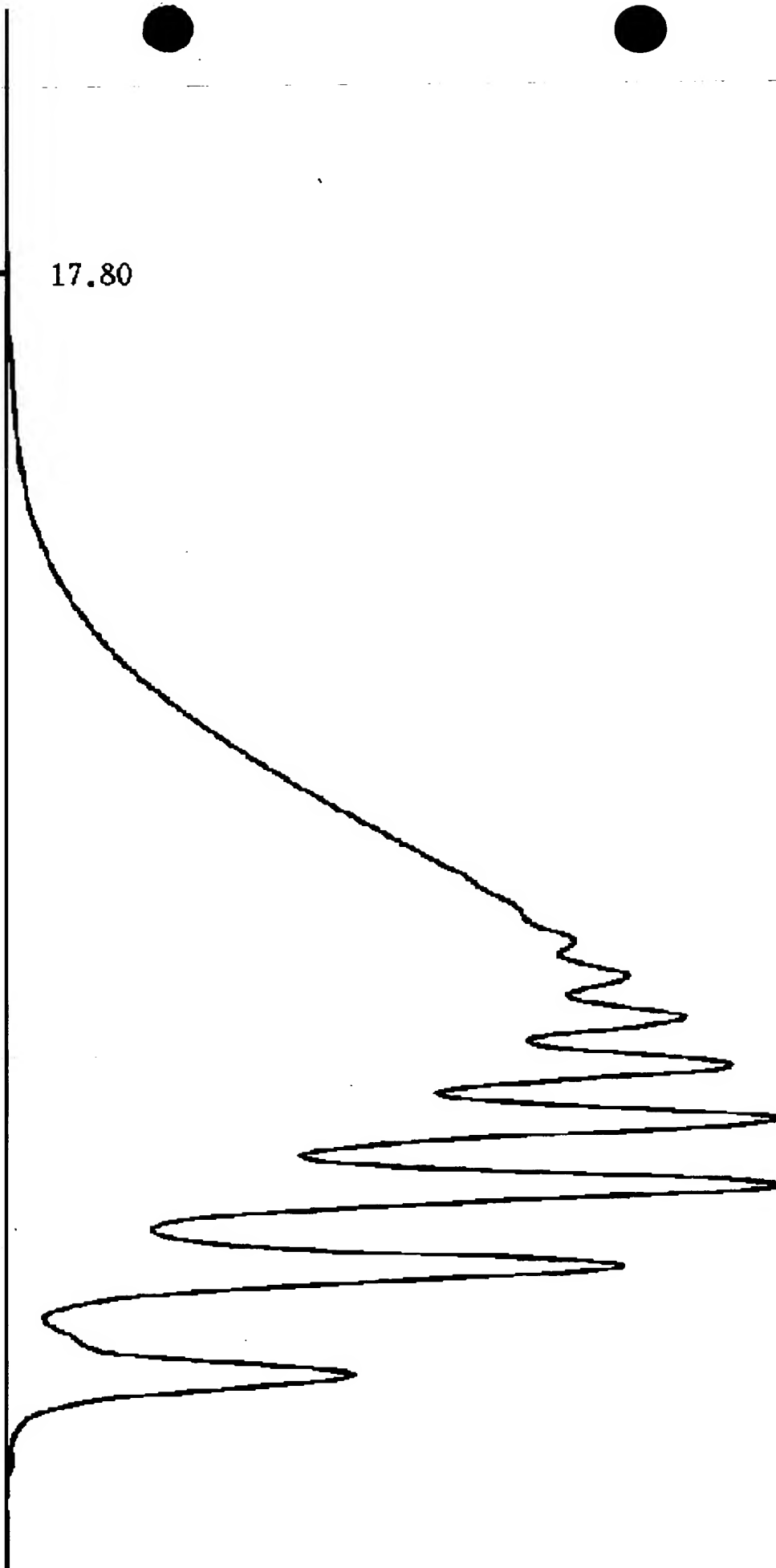


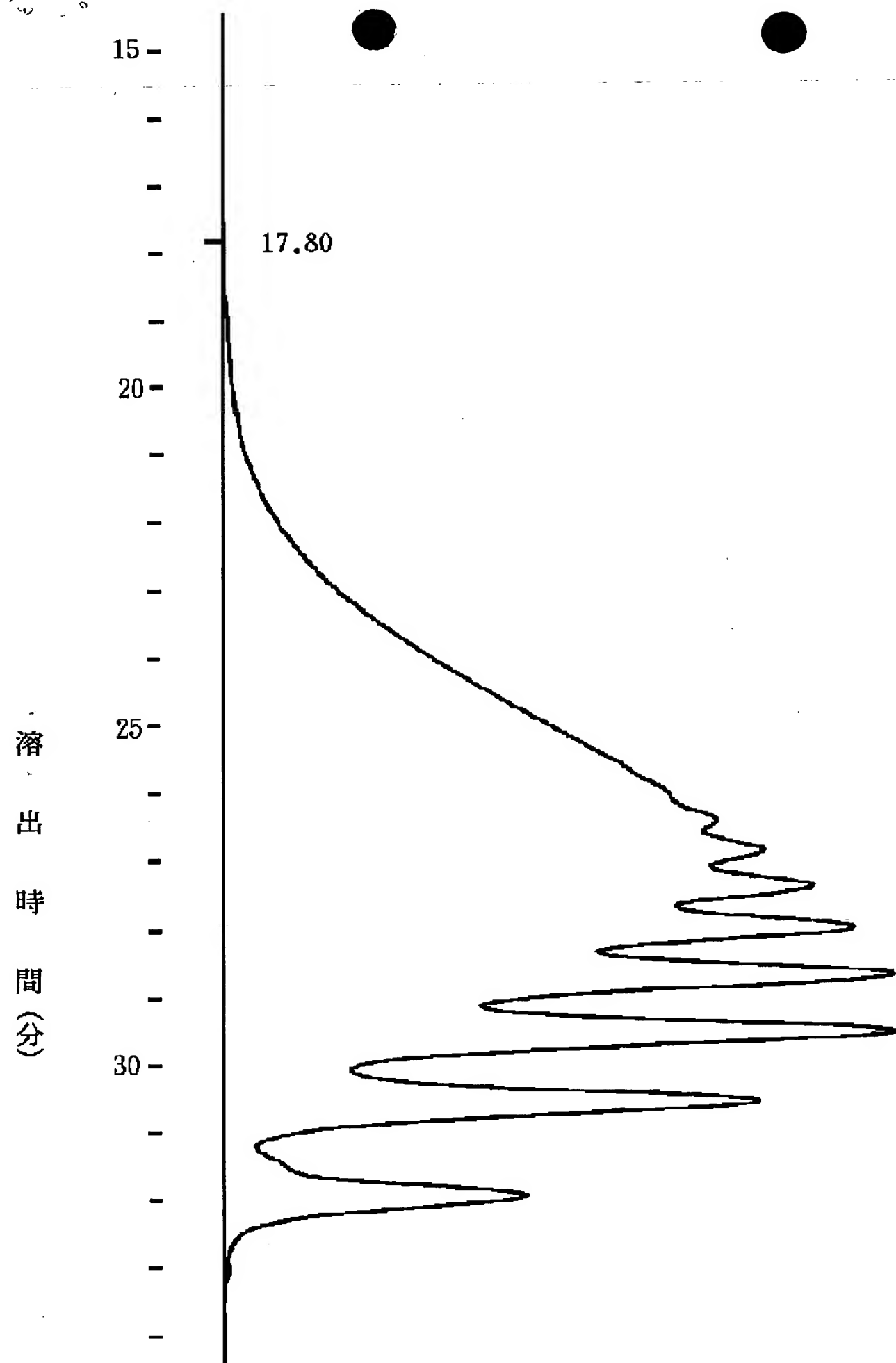


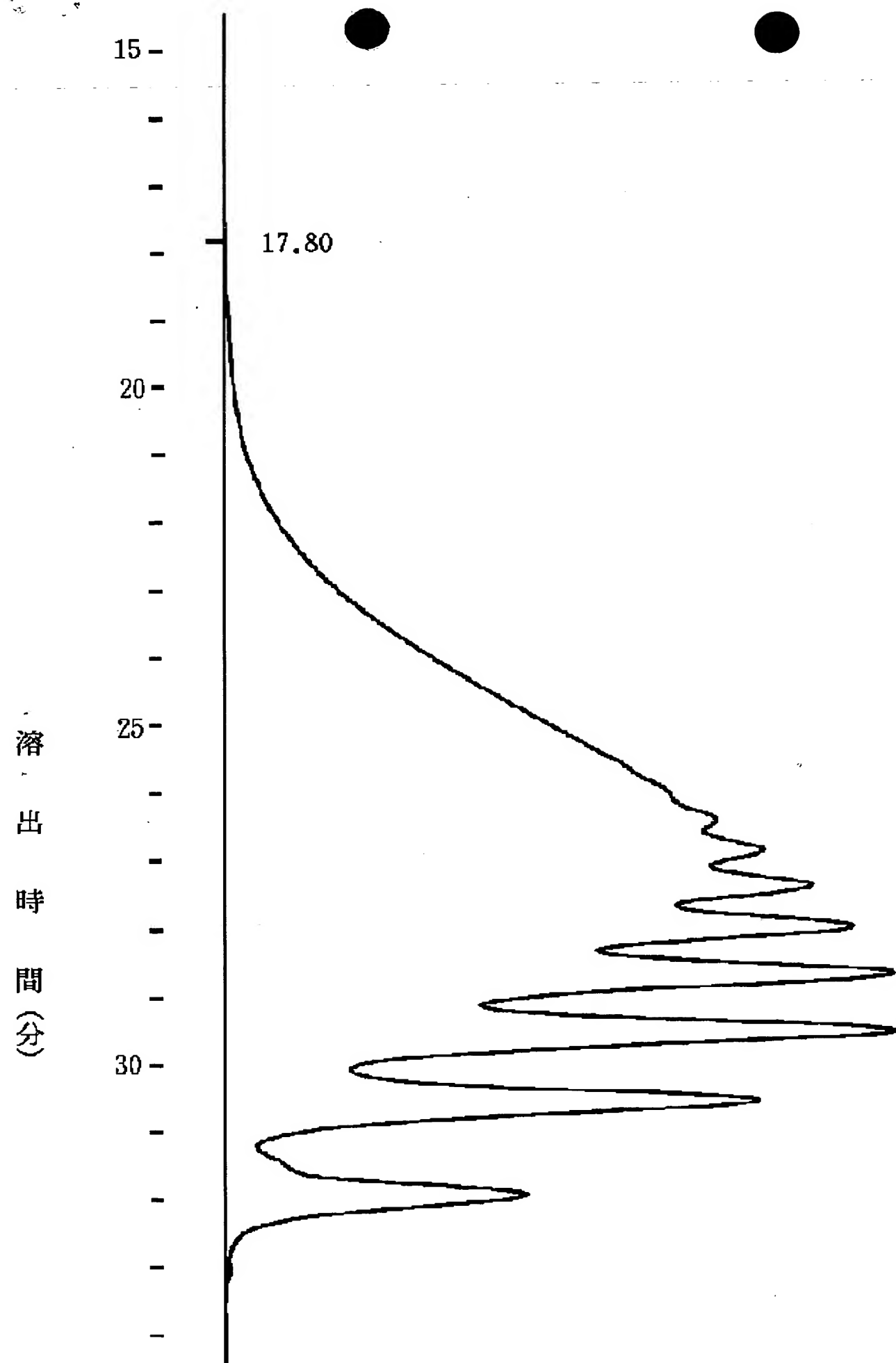
溶出時間(分)

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